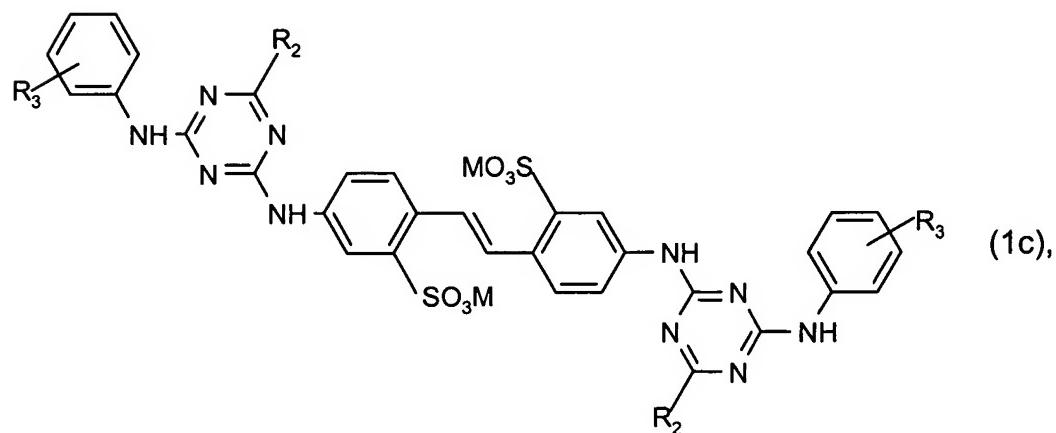
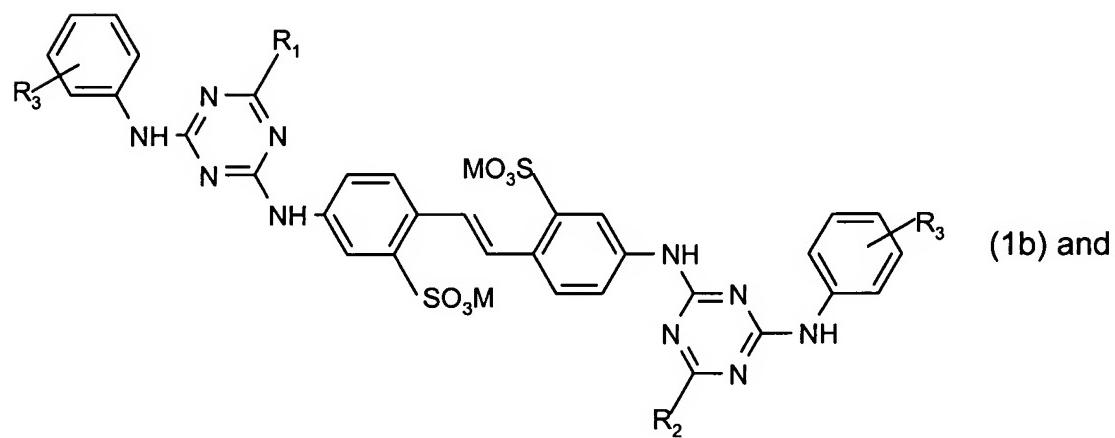
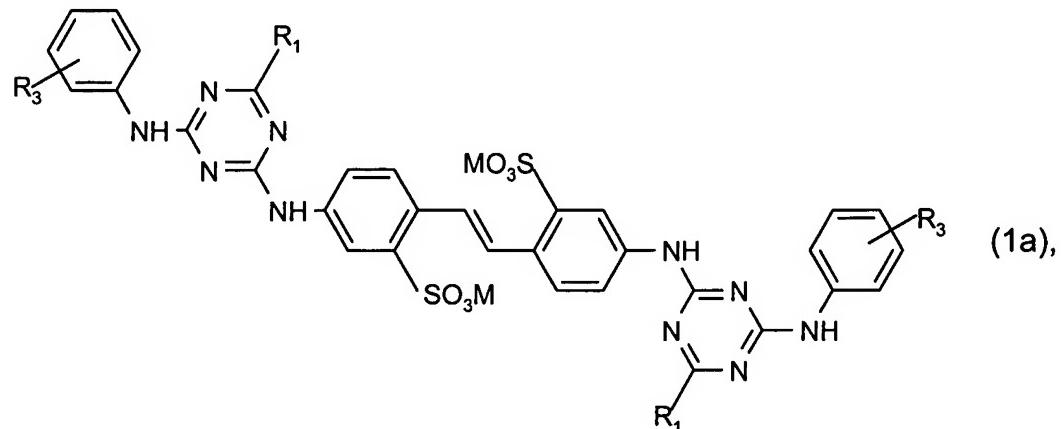


Claims

1. (previously presented) A fluorescent whitening agent, which comprises a mixture of two symmetrical compounds 1(a) and 1(c) and one asymmetrical compound 1(b) of the formulae



in which R_1 and R_2 are different and

R_1 represents $-NH_2$, $-NHC_1-C_4alkyl$, $-N(C_1-C_4alkyl)_2$, $-NHC_2-C_4$ hydroxyalkyl, $-N(C_2-C_4hydroxyalkyl)_2$, $-N(C_1-C_4alkyl)(C_2-C_4$ hydroxyalkyl), a morpholino, piperidino or pyrrolidino residue,

R_2 represents $-NH_2$, $-NHC_1-C_4alkyl$, $-N(C_1-C_4alkyl)_2$, $-NHC_2-C_4$ hydroxyalkyl, $-N(C_2-C_4hydroxyalkyl)_2$, $-N(C_1-C_4alkyl)(C_2-C_4$ hydroxyalkyl), a morpholino, piperidino or pyrrolidino residue or an amino acid or an amino acid amide residue from which a hydrogen has been removed from the amino group,

each R_3 , independently, represents hydrogen, C_1-C_4alkyl or $C_1-C_4alkoxy$ and

M represents hydrogen, an alkali metal atom, ammonium or a cation formed from an amine.

2. (original) A composition according to claim 1, in which R_3 represents hydrogen.

3. (previously presented) A composition according to claims 1, in which the aliphatic amino acid or amino acid amide residue is of the formula

$-NR_4-CH(CO_2H)-R_4$ (2) or $-NR_4-CH_2CH_2CONH_2$ (3),

in which each

R_4 and R_4 , independently, represent hydrogen or a group having the formula

$-CHR_5R_6$ in which

R_5 and R_6 , independently, are hydrogen or C_1-C_4alkyl optionally substituted by one or two substituents selected from the group consisting of hydroxy, thio, methylthio, amino, carboxy, sulfo, phenyl, 4-hydroxyphenyl, 3,5-diiodo-4-hydroxyphenyl, β -indolyl, β -imidazolyl and $NH=C(NH_2)NH-$.

4. (previously presented) A composition according to claim 3, in which residues R_2 are derived from glycine, alanine, sarcosine, serine, cysteine, phenylalanine, tyrosine (4-hydroxyphenylalanine), diiodotyrosine, tryptophan (β -indolylalanine), histidine ((β -imidazolylalanine), α -aminobutyric acid, methionine, valine (α -aminoisovaleric acid), norvaline, leucine (α -aminoisocaproic acid), isoleucine (α -amino- β -methylvaleric acid), norleucine (α -amino-n-caproic acid), arginine, ornithine (α , δ -diaminovaleic acid), lysine (α , ϵ -diaminocaproic acid), aspartic acid (aminosuccinic acid), glutamic acid (α -aminoglutaric acid), threonine, hydroxyglutamic acid and taurine, as well as mixtures and optical isomers thereof, or from iminodiacetic acid or from N-(propionamido)-N-(2-hydroxyethyl)amine.

5. (previously presented) A composition according to claim 1, in which R₂ represents -NHC₂-C₄hydroxyalkyl, -N(C₂-C₄ hydroxyalkyl)₂, -N(C₁-C₄alkyl)(C₂- C₄hydroxyalkyl), a morpholino residue or a residue derived from glycine, sarcosine, taurine, glutamic acid, aspartic acid or iminodiacetic acid.

6. (previously presented) A composition according to claim 5 in which R₂ represents a mono-(2-hydroxyethyl)amino, a di-(2-hydroxyethyl)amino, a di-(2-hydroxypropyl)amino, an N-(2-hydroxyethyl)-N-methylamino, an aspartic acid, an iminodiacetic acid or a morpholino residue.

7. (previously presented) A composition according to claim 1, in which M represents hydrogen, lithium, potassium, sodium, ammonium, mono-, di-, tri- or tetra-C₁-C₄alkylammonium, mono-, di- or tri-C₁-C₄hydroxyalkylammonium or ammonium that is di- or tri-substituted with a mixture of C₁-C₄alkyl and C₁-C₄hydroxyalkyl groups.

8. (original) A composition according to claim 7, in which M represents hydrogen, potassium or sodium.

9. (previously presented) A process for the preparation of the compound mixture of formulae (1a), (1b) and (1c) of claim 1 which process comprises reacting, under known reaction conditions, cyanuric chloride, successively, in any desired sequence, with each of

- i) 4,4'-diaminostilbene-2,2'-disulphonic acid,
- ii) aniline or aniline substituted by C₁-C₄alkyl or C₁-C₄alkoxy,
- iii) an amino compound R₁H and
- iv) an amino compound R₂H

or, alternatively

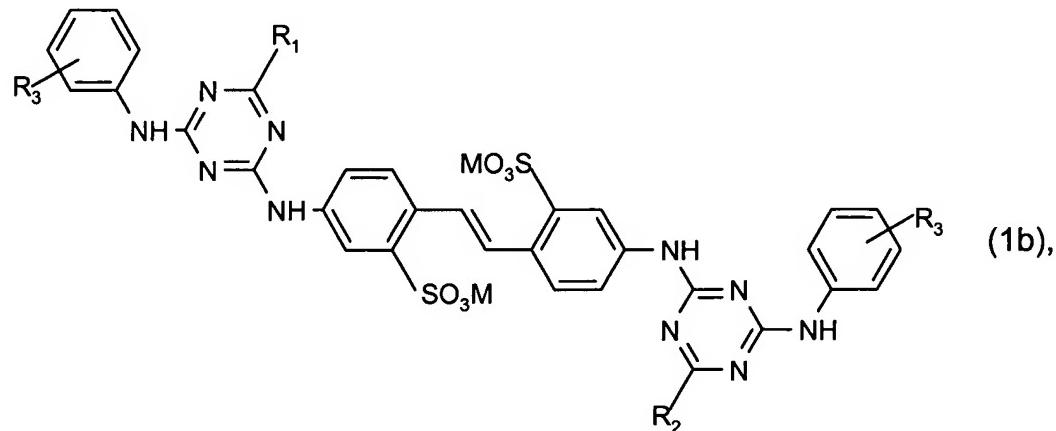
- i) 4,4'-diaminostilbene-2,2'-disulphonic acid,
- ii) aniline or aniline substituted by C₁-C₄alkyl or C₁-C₄alkoxy, and
- iii) a mixture of an amino compound R₁H and an amino compound R₂H

wherein R₁ and R₂ are different and

R₁ represents -NH₂, -NHC₁-C₄alkyl, -N(C₁-C₄alkyl)₂, -NHC₂-C₄ hydroxyalkyl, -N(C₂-C₄hydroxyalkyl)₂, -N(C₁-C₄alkyl)(C₂-C₄ hydroxyalkyl), a morpholino, piperidino or pyrrolidino residue and

R_2 represents $-NH_2$, $-NHC_1-C_4alkyl$, $-N(C_1-C_4alkyl)_2$, $-NHC_2-C_4$ hydroxyalkyl, $-N(C_2-C_4hydroxyalkyl)_2$, $-N(C_1-C_4alkyl)(C_2-C_4$ hydroxyalkyl), a morpholino, piperidino or pyrrolidino residue or an amino acid or an amino acid amide residue from which a hydrogen has been removed from the amino group.

10. (previously presented) A compound of the formula



in which

R_1 and R_2 are different and

R_1 represents $-NH_2$, $-NHC_1-C_4alkyl$, $-N(C_1-C_4alkyl)_2$, $-NHC_2-C_4$ hydroxyalkyl, $-N(C_2-C_4hydroxyalkyl)_2$, $-N(C_1-C_4alkyl)(C_2-C_4$ hydroxyalkyl), a morpholino, piperidino or pyrrolidino residue,

R_2 represents $-NH_2$, $-NHC_1-C_4alkyl$, $-N(C_1-C_4alkyl)_2$, $-NHC_2-C_4$ hydroxyalkyl, $-N(C_2-C_4hydroxyalkyl)_2$, $-N(C_1-C_4alkyl)(C_2-C_4$ hydroxyalkyl), a morpholino, piperidino or pyrrolidino residue or an amino acid or an amino acid amide residue from which a hydrogen has been removed from the amino group,

R_3 represents hydrogen, C_1-C_4alkyl or $C_1-C_4alkoxy$ and

M represents hydrogen, an alkali metal atom, ammonium or a cation formed from an amine.

11. (previously presented) A composition for whitening synthetic or natural organic materials, which composition contains water, a fluorescent whitening agent comprising a mixture of the compounds (1a), (1b) and (1c), according to claim 1, and, optionally, one or more auxiliaries selected from the group consisting of dispersants, water retention aids, biocides and adjuvants.

12. (previously presented) A method for adding optical brightening agents to paper which method comprises the step of applying a composition of claim 11 either to a paper substrate in a pulp mass, to a paper substrate in a size-press, to a paper substrate in a metering press or contacting a paper surface with a coating application comprising a composition of claim 11.

13. (cancelled)

14. (previously presented) A method, for increasing the Sun Protection Factor (SPF) rating or for the fluorescent whitening of a textile fibre material which method comprises the step of treating said textile fibre material with a composition of claim 11.

15. (cancelled)

16. (previously presented) A process according to claim 9, wherein cyanuric chloride is initially reacted with 4,4'-diaminostilbene-2,2'-disulphonic acid.

17. (previously presented) A process according to claim 16, wherein cyanuric chloride is initially reacted with 4,4'-diaminostilbene-2,2'-disulphonic acid, followed by reaction with aniline or aniline substituted by C₁-C₄alkyl or C₁-C₄alkoxy and then with a mixture of amino compounds R₁H and R₂H.